Interactions between solidification and compositional convection in alloys

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Abstract

This project combined theoretical and experimental ground-based studies of the interactions between convection and solidification of binary melts. Particular attention was focused on the alteration of the composition and microstructure of castings caused by convective flows through the interstices of mushy layers. Two different mechanisms causing convection were investigated. (i) Compositional, buoyancy driven convection is known to cause chimneys and freckles in directionally cast alloys on Earth. The analytical studies provide quantitative criteria for the formation of chimneys that can be used to assess the expediency of producing alloys in Space. (ii) Flow of the melt is also driven by the contraction (expansion) that typically occurs during change of phase. Such convection will occur even in the absence of gravity, and may indeed be the primary cause of macrosegregation during the production of alloys in Space.

The studies will employed asymptotic methods in order to determine conditions for the stability of various states of solidifying systems. Further, simple macroscopic models of complete systems were developed and solved. These analytical studies were augmented by laboratory experiments using aqueous solutions, in which the convective flows could be easily observed and the effects of convection could be readily measured. These experiments guided the development of the theoretical models and provided data against which the predictions of the models can be tested.

Achievements

Chiareli completed a thorough, fundamental study of the flow through mushy layers driven by expansion during solidification. Such convection is an inevitable consequence of solidification and cannot be avoided by manufacturing materials in space. It causes macrosegregation in a casting, which was analysed with a set of similarity solutions of equations developed to describe the interstitial flow in a mushy layer driven by expansion.

The flow driven by expansion (in silicon-based products, for example) can drive an instability that leads to the flow becoming focused and the casting becoming laterally inhomogeneous. The potential for instability was proved, but it was found that the instability is unlikely to occur under typical casting conditions.

Worster (in collaboration with RC Kerr from the Australian National University) conducted a suite of experiments in which the degree of macro-segregation caused by compositional (buoyancy-driven) convection was quantified. A particular focus of this study was the observation that adding small quantities of an impurity to the melt (in the experiments, copper sulphate was added to aqueous solutions of ammonium chloride) could inhibit the formation of chimneys (freckles) that formed in the casting. A theoretical model was developed that showed how supercooling at the mush–liquid interface enhances a particular mode of convection in the melt that influences the state of the underlying mushy layer in such a way as to suppress convection within the mushy layer and hence to suppress the occurrence of chimneys. Experimental measurements showed that the addition of impurities can increase the degree of supercooling significantly. The predictions of the theoretical model gave good agreement with the experimental observations of the time of chimney formation and the amount of impurity needed to suppress the formation of chimneys.

Anderson carried out a significant nonlinear analysis of convection in mushy layers and thereby determined the physical mechanisms that control the degree of sub-criticality of the bifurcations to convection. The study yielded a comprehensive set of coupled amplitude equations, which were used to predict the modes of convection in a mushy layer and their stability.

In the course of this nonlinear study, Anderson discovered a hitherto unsuspected oscillatory mode of instability in the mushy layer that is a consequence of the intimate coupling between solidification and convection and may cause compositional striations to occur in a cast alloy. This instability (which is a linear phenomenon) was studied in some detail as it elucidated some of the complex dynamics that can occur in mushy layers.

Schulze developed a sophisticated numerical model of convection in a mushy layer with fully-developed chimneys. Although the chimneys were not fully resolved, their gross properties (in particular their width and the mass flux through them) were allowed to vary dynamically in response to the buoyancy-driven flow, as was the height of the mushy layer. The numerical study was supported by a theoretical analysis of the various boundary layers that form and a scaling analysis of the gross features of the layer and flow within it. These studies have extended Anderson's weakly nonlinear results to a fully nonlinear regime and showed that chimneys can form nonlinearly at even smaller Rayleigh numbers than predicted by an analysis of nonlinear convection without chimneys.

Worster (in collaboration with JS Wettlaufer, University of Washington, and HE Huppert, University of Cambridge) performed extensive laboratory experiments to study the formation and evolution of sea ice. The data from these experiments is being used to guide the development of predictive mathematical models that will aid the remote sensing by satellite of the freezing of the Arctic oceans, which have a major effect on the changing global climate. Though focused on sea ice, this study has provided quantitative data confirming predictions for the onset of convection in mushy layers generally made by Worster, Anderson and Schulze previously in this project.

Though not originally proposed, Worster additionally engaged in a fundamental study (with Wettlaufer) of solidification in layers premelted liquid layers a few tens of molecular diameters thick. This is leading to an understanding of the transport of material in partially-molten systems, such as frozen ground and sintered materials. Such transport can far exceed any gravitational flow and is responsible for a number of defects in materials grown in micro gravity.

Publications resulting from project

Segregation and Flow during the Solidification of Alloys (1994) *Journal of Crystal Growth* 139, 134–146 (with A.O.P. Chiareli and H.E. Huppert).

The Transient Behaviour of Alloys Solidified from Below Prior to the Formation of Chimneys (1994) *Journal of Fluid Mechanics* 269, 23–44 (with R.C. Kerr).

The dynamics of premelted films: frost heave in a capillary (1995) *Phys. Rev.* E 51, 4679 (with J.S. Wettlaufer).

Flow focusing instability in a solidifying mushy layer. (1995) *Journal of Fluid Mechanics* 297, 293–305 (with A.O.P. Chiareli).

Weakly-nonlinear analysis of convection in a mushy layer during the solidification of alloys. (1995) *Journal of Fluid Mechanics* 302, 307–331 (with D.M. Anderson).

A new oscillatory instability in a mushy layer during the solidification of binary alloys. (1996) *Journal of Fluid Mechanics* 307, 245–267 (with D.M. Anderson).

A theory of premelting dynamics for all power law forces. (1996) *Phys. Rev. Lett.* 76, 3602 (with J.S. Wettlaufer, L.A. Wilen & J.G. Dash).

Dynamic contact angles in containerless solidification. (1996) *Journal of Crystal Growth* 163/3, 329–338 (with D.M. Anderson & S.H. Davis).

Convection in mushy layers. (1997) *Ann. Rev. Fluid Mech.* 29, 91–122

The phase evolution of young sea ice. (1997) *J. Geophys. Res.* Geophys. Res. Lett. 23, 1251-54 (with J.S. Wettlaufer & H.E Huppert)

An experimental study of the formation and evolution of sea ice. (1997) *Journal of Fluid Mechanics* 344, 291–316 (with J.S. Wettlaufer & H.E Huppert)

Natural convection, solute trapping and channel formation during solidification of salt water. (1997) *Journal of Physical Chemistry* 101 (32), 6132–36 (with J.S. Wetlaufer)

Premelting Dynamics: Geometry and Interactions. (1997) *Journal of Physical Chemistry* 101 (32), 6132–36 (with J.S. Wetlaufer & L.A. Wilen)

A numerical investigation of steady convection in mushy layers during the directional solidification of binary alloys (1998) *Journal of Fluid Mechanics* 356, 199–220 (with T.P. Schulze)